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[54] Patent Title: **Reactor, for Use in Carrying Out Endothermic or Exothermic Catalytic Reactions**

PATENT CLAIMS

Claim 1) A catalytic reactor, for use in carrying out endothermic or exothermic reactions, through which a reactant fluid is flowed, and containing a reaction chamber filled with catalyst material, which is in thermal contact with a heat-emitting or heat-absorbing fluid, and characterized by the fact that the cross-section surface area of the reaction chamber is varied, along the direction of flow of the reacting fluid, depending upon the quantity of heat required for completion of a given reaction, or the quantity of heat released on the course of a reaction.

Claim 2) A catalytic reactor of the type described above, in Claim 1, further characterized by the fact that the reaction chamber is composed essentially of pipes [5, 13, 14, 15], which have variable cross-section diameters.

Claim 3) A catalytic reactor of the type described above, in Claim 1, further characterized by the fact that the reaction chamber is composed essentially of pipes [12], said pipes [12] exhibiting a constantly changing diameter along the direction of flow.

Claim 4) A catalytic reactor of the type described above, in Claim 1, further characterized by the fact that the reaction chamber contains catalyst material, into which sleeves [17] are inserted, which possess varying diameters along the direction of flow of the reaction fluid, and which contain tubes [16] through which the heat-transfer fluid circulates.

Claim 5) A catalytic reactor of the type described above, in Claims 1 through 4, further characterized by the fact that a portion of the reaction chamber contains a transverse section, which is divergent from the rest of the reaction chamber, and which is oriented along the direction of flow of the reaction fluid. This divergent section can be directly connected to the reaction fluid inflow line, and can operate either alternatively to, or in conjunction with, the remaining processing volume of the reactor.

Claim 6) A catalytic reactor of the type described above, in one of the following Claims - 2, 3 or 5, further characterized by the fact that at least two of the adjacent pipes [5] are positioned antiparallel to the direction of fluid flow.

Claim 7) A catalytic reactor of the type described above, in one of the following Claims - 2, 3, 5 or 6, further characterized by the fact that at least two of the pipes [13, 14] within the reactor are connected to a larger tube [15].

Claim 8) A catalytic reactor of the type described above, in one of the following Claims - 2, 3, 5, 6 or 7, further characterized by the fact that, for any given pipe [5, 12, 13, 14, 15] diameter, D_i (in mm), the relation which prevails for the quantity of gas flowing through a pipe of the specified diameter (D_i), per unit time, V (in Nm/h), is as follows:

$$D_i = p \cdot V^q,$$

where 'p' and 'q' are constants.

DETAILED DESCRIPTION OF INVENTION

(Reactor for Use in the Carrying Out of Endothermic or Exothermic Reactions)

The present invention comprises A catalytic reactor, for use in carrying out endothermic or exothermic reactions, through which a reactant fluid is flowed, and containing a reaction chamber filled with catalyst material, which is in thermal contact with a heat-emitting or heat-absorbing fluid.

At the present time, there are many kinetic reaction systems - such as, for instance, those employed in the synthesis of methanol or ammonia, which are operated in the presence of catalytic substances. In accordance with the prevailing operational and reaction conditions in a given case (catalyst materials and geometric relations within the reactor), a gas composition is established, in the course of the reaction, which is more or less close to the equilibrium composition. In many cases of this type, several reactions are occurring simultaneously within the reactor, each reaction having its own specific rate and enthalpy value. During such procedures, numerous temperature regions are created within the reactor, as a result of the temperature of the catalyst material employed and/or owing to the generation, from below, of undesirable byproduct substances, as a result both of a periodically desirable approximation to the equilibrium condition, and of a the economic and limitations imposed by the economic and material constraints, including the catalyst material. In any case, the production of the desired temperature gradient within the reactor is an important precondition for the successful completion of a given reaction procedure.

In the journal "Chemtech" (July, 1973: pp. 430 - 435), there is a description of a methanol reactor of the type described above, which has catalyst material placed inside the reaction pipes. A reactant gas is flowed through these pipes. The heat generated in the course of the synthesis reaction is carried off by cold water (cooling fluid), which is flowed around the outside of the reaction pipes. In the example provided in the cited

article, the desired temperature profile is obtained on the basis of a gas inflow temperature which is lower than the temperature of the cooling fluid; in the course of the catalytic reaction, the gas temperature is raised to a value ca. 11° C above that of the cooling fluid, and, conditioned by the reduction of both heat of reaction heat transfer to the cooling fluid, there is an associated cooling to the gas outflow temperature, which is a few °C above the temperature of the cooling fluid.

In the reactor under discussion, heat from the gas and catalyst material is initially transferred to the cooling fluid. In cases in which a higher initial gas temperature is selected, the highest temperature zone within the reactor will be displaced upward, which, however, is practicable only to a limited extent owing to the potential for damaging the catalyst material, the prevailing equilibrium conditions, and the impact on process cost-effectiveness.

The present invention provides the advantage of comprising a reactor of the general type in question here, which is characterized by an exceptionally good adjustment of the actual reactor temperature profile to the most favorable values for the completion of the reaction in question. The proposed reactor is relatively simple and inexpensive to install and operate.

In the proposed reactor, the problems discussed above, in conjunction with the description of existing reactors of the same general type, are resolved in the following manner: the cross-section area of the reaction chamber is varied, along the direction of flow of the reactant fluid, in accordance with the specific quantity of heat which must be contributed to, or removed from, the reaction zone.

The cross-section area of the reaction chamber is adjusted to provide a theoretically optimal temperature gradient for the system: in cases in which large quantities of heat are released (in exothermic reactions) or required (in endothermic reactions), a relatively small diameter reaction chamber (tubing) cross-section area is employed; while, conversely, if the quantities of heat which must be removed from or added to the system are relatively small, tubing of a correspondingly large diameter and cross-section area is employed. In the first instance, there is a relatively high ratio of heat-supplying or -removing fluid in contact with the tube wall surfaces, in relation to total catalyst volume within the tubing, and, as a result, the system possesses a markedly high value for heat-transfer capability to or from the catalyst material. In the second scenario, the above ratio is relatively low, and, as a result, only a quite limited amount of heat-transfer activity is available.

The present invention employs a surprisingly simple solution to provide the optimal temperature gradient for the reactor. As a result of variations in tubing diameter of the reaction chamber, areas in which relatively large or small quantities of heat are produced, or required, can be provided with optimal reaction conditions.

The present invention is suitable for use in both single-phase (gaseous reaction partner) and biphase (gaseous and liquid reaction partners) system, and with either fixed--bed or flow-bed catalyst installations.

In the proposed reactor, the authors have found it most advantageous to employ a reaction area which consists primarily of tubing, with the tubing possessing varying cross-section diameters. In this configuration, in which the catalyst material is placed inside the reaction tubes, the tubes are positioned parallel to the flow direction of the reactant fluid. The reaction tubes are surrounded by a fluid which either adds heat to the system, or removes heat from the system, in order to facilitate, respectively, an endothermic or an exothermic reaction. Control over reactor conditions can be enhanced, in the context of the present invention, through the use of reaction tubes which not only possess varying diameters and cross-section areas, but which are also of varying lengths.

In another advantageous form of the present invention, wherein the reaction area also consists primarily of tubing, the reaction tubes exhibit a continuously changing diameter along the direction of flow of the reacting fluid.

In another advantageous form of the present invention, sleeves of continuously varying diameter, along the flow direction of the reacting fluid, are inserted into the catalyst material, and, within these sleeves, there are tubes which function in the supplying or removal of the requisite quantities of heat. The cross-section are for fluid flow is more or less constricted in accordance with the disposition of the sleeves. In this variant of the present invention, the catalyst material within the sleeves is penetrated by the heat-transfer tubes. In this system, the constriction of the flow stream of the reacting fluid results in a corresponding increase in the ratio of surface area to reaction chamber catalyst volume, while increasing the flow stream diameter has the opposite effect. It is clear that the widening or narrowing of the reacting fluid flow stream by the sleeves can be achieved through the use of either stepwise or continuous variation of the cross-section area.

In cases in which the reactor is either over- or undercharged, system equilibrium must be established through the use of non-standard procedures, since there are corresponding changes in liquid flow rates, reaction enthalpy values, and quantity of converted liquid. Even in cases in which a reactor has been optimized for use under normal operating conditions, an under- or overcharge situation can generate less than optimal conditions within the reactor. For this reasons, the authors of the present invention have incorporated into the proposed reactor a portion of the reaction area should contain a transverse section, diverging from the remainder of the reaction area, and positioned along the flow direction of the reacting fluid, and which can be connected to the reacting fluid inflow tube in order to permit it to operate, either as an alternative or to or a supplement for, the remaining reaction area.

In this latter variant of the proposed reactor, one section of the reaction area contains a transverse section, which is positioned along the flow direction of the reacting fluid, and

which is adapted in a manner which provides optimal conditions for normal operation. There is, in addition, a reaction zone which is optimized for use in under- or overcharge conditions, and which can be used, under such non-standard conditions, either in conjunction with, or instead of, the other reactor section. Thus, optimized reactor operation can be maintained under a wide range of load conditions.

In another advantageous variant of the present invention, adjacent reaction tubes are positioned antiparallel to the flow direction of the reacting fluid. This configuration is advantageous in cases in which, [finish sentence].
As a result, the volume of the reaction area can be substantially reduced.

In a modification of the present invention, at least two of the reaction tubes within the reactor are combined to form a single tube.

In the proposed reactor, for any given reaction tube, the diameter, D_i (in mm), the relation which prevails for the quantity of gas flowing through a pipe of the specified diameter (D_i), per unit time, V (in Nm/h), is as follows:

$$D_i = p \cdot V^q,$$

where 'p' and 'q' are constants. Typical values ranges for 'p' and 'q' for a methanol synthesis reactor might be as follows: $15 \leq p \leq 25$; and $0.12 \leq q \leq 0.22$.

The proposed reactor is particularly well suited for use in ammonia synthesis or methanol synthesis operations, but the potential scope of its application is not necessarily limited to these procedures.

A more detailed representation of the present invention is provided by the schematic diagrams, below.

Various forms of the proposed reactor are provided below, in Figs. #1 through #6.

In all of these diagrams, [1] represents the reactor, which is situated in a casing [2]. The proposed reactor [1] is constructed in the manner of a tubular heat exchanger, and contains two separate flow areas.

In Figs. #1 through #5, the flow area [4] comprises several tubes [5], which are positioned within the casing [2], and are attached to tube plates [6] at each end. Both sides of the reaction tubes are connected with collection zones [7a & 7b], which, in turn, are connected, respectively, to the inflow [8] and outflow [9] pipelines. The remaining flow area [3], which is bounded by the inner walls of the casing [2], contains a heat-supplying or -removing fluid, which is added to and removed from the reactor, respectively, through a lower inflow aperture [10] and an upper outflow aperture [11]. The reaction tubes [5] are filled with a suitable catalyst material. In the context of the present invention, the diameters of the reaction tubes [5] are varied along the flow direction of the reacting fluid, with diameters being selected in such a manner that the

best possible equilibrium and/or heat-exchange conditions are created and maintained within the system. In areas where large quantities of heat are generated or required (in accordance, respectively, with whether the reaction in question is exothermic or endothermic in nature), the reaction tube (wider tubes; higher ratio value) has a relatively small-diameter and cross-section area, in order to decrease the ratio of reaction volume to surface area in contact with the heat-transfer fluid; while the opposite is the case for areas which generate or require relatively small quantities of heat.

The reactor represented below, in Fig. #1, is well-suited to provide a selected temperature profile within the system.

The reactor depicted in Fig. #2, which might be utilized, for instance, in a methanol synthesis operation, exhibits the following representative parameter values:

. Gas composition upon entry into the reactor:	17% CO; 73 H_2; 5 CO_2; 5% inert.
. Gas flow rate per reaction tube:	V = 200 Nm^3/h.
. Gas entry temperature:	245° C.
. Cooling water entry temperature:	230° C.
. First tube cross-section diameter:	45 mm.
. Second tube cross-section diameter:	52 mm.
. Third tube cross-section diameter:	45 mm.
. Length of first tube cross-section zone:	2.5 m.
. Length of second tube cross-section zone:	1.5 m.
. Length of third tube cross-section zone:	4.0 m.
. Spatial rate for the first tubing zone:	40,000 - 50,000 V/V_{cat}.
. Spatial rate for the second tubing zone:	55,000 - 70,000 V/V_{cat}.
. Spatial rate for the third tubing zone:	30,000 - 40,000 V/V_{cat}.

[V_{cat} = volume of catalyst material, in m^3].

In more general terms, for reactors of the type represented in Fig. #2, the typical range of reaction tubing cross-section zone lengths is from 2 to 5 (for instance, 3), and the typical range for tubing cross-section diameter is from 20 to 100. In such cases, typically, the conditions for diameter (D_1, in mm) and gas flow per hour (V; in Nm^3/h) would be as follows:

$$D_1 = (15 \ldots 25) \cdot V^{0.12} \ldots {}^{0.22}.$$

In Fig. #3, below, there is a diagrammatic representation of a reactor in which adjacent reaction tubes are positioned antiparallel to the reaction fluid direction of flow. A configuration of this type is well suited for use in tubing sections with relatively large cross-section areas are positioned adjacent to sections with relatively small cross-section areas. In this manner, the size of the reaction chamber, and of the reactor itself, can be substantially reduced.

Fig. #4 depicts a reactor in which the diameters of the reaction tubes [5] vary continuously along the direction of flow of the reaction fluid.

In Fig. #5, below, there is a schematic representation of a reactor in which two reaction tubes join to forma single common tube. A structural configuration of this type provides additional variation possibilities, for use in the simultaneous optimization of reaction equilibrium and reaction volume.

Fig. #6 represents a reactor with a modified structural configuration. In this case, the catalyst material is placed, not inside the reaction tubes, but, instead, within the entire inner volume of the reactor, limited by the inner walls of the casing [2]. Here, the heat-transfer fluid flows through the tubes [16] which are themselves positioned within the sleeves [17]. The reacting fluid enters through an aperture situated on the top of the reactor [8], and is removed through an aperture [9] on the bottom of the reactor.
The sleeves [17] are inserted into the catalyst material and limit the lateral expansion of the flow stream of reaction fluid. By this means, the cross-section area of said flow stream can be widened or narrowed, as prevailing conditions require. In this variant, the changing of flow stream cross-section area can be carried out in either a stepwise or a continuous manner.

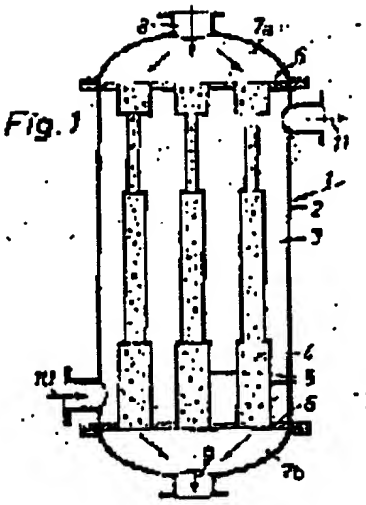

Fig. 1

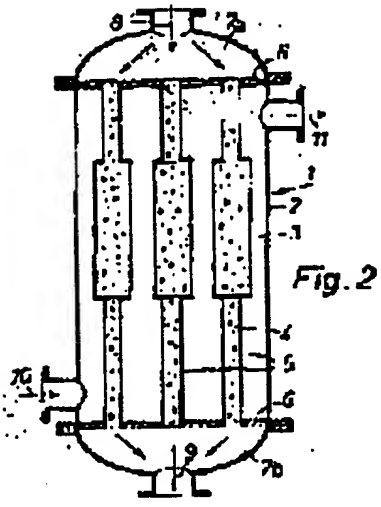

Fig. 2

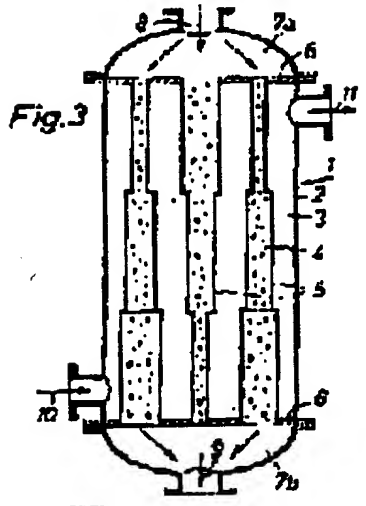

Fig. 3

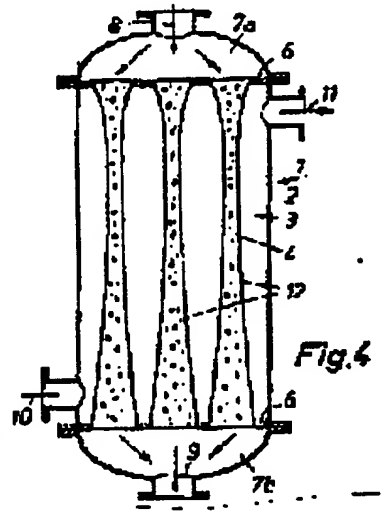

Fig. 4

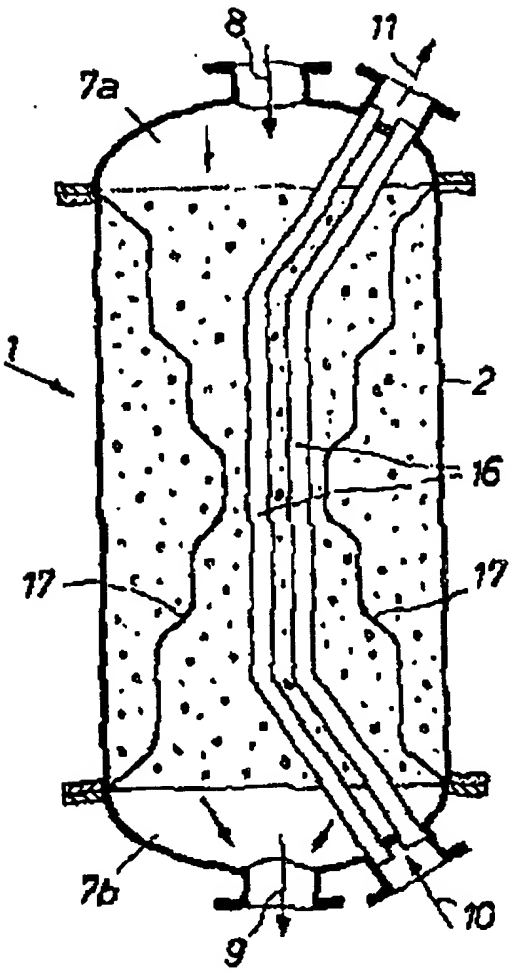

Fig.6

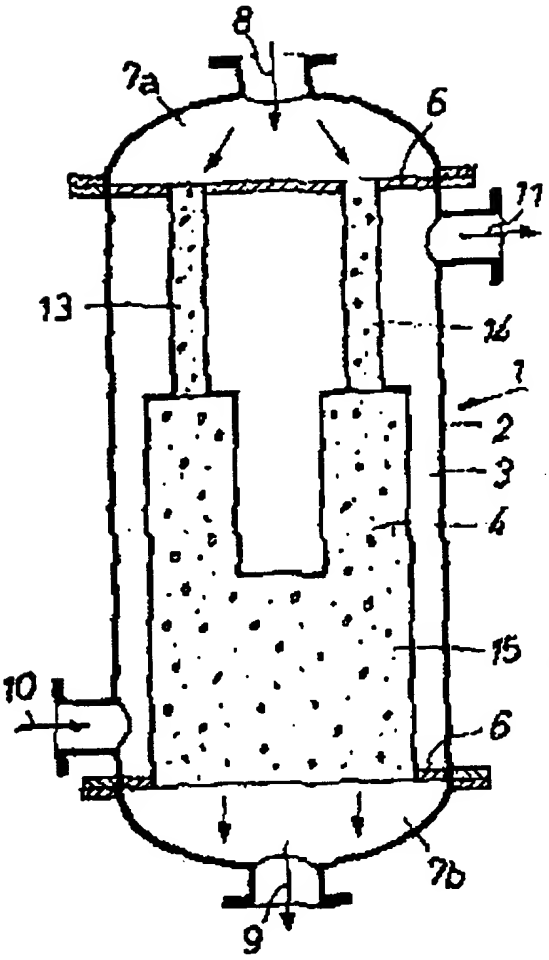

Fig. 5